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Journal of the European Ceramic Society 26 (2006) 683–687
www.elsevier.com/locate/jeurceramsoc

Observation of the granule packing structure using a confocal laser-scanning microscope

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Available online 24 August 2005

Abstract

A new method was developed to examine the change in internal structure of a granule compact for the initial stage of compaction. Using a resin of high refractive index mixed with a fluorescent reagent, the internal structure of alumina granule compact was clearly observed three dimensionally by a confocal laser-scanning microscope (CLSM). This method has a high potential to investigate the granule deformation process in the initial stage.

Keyword: Alumina granule compact; Pressing; Mechanical properties

1. Introduction

Granule compaction is the most common forming method for fabricating ceramics green compacts. Detrimental defects are often formed in a green body with this method during the forming process, reducing the reliability of ceramics after sintering.[1] Full characterization of these defects is very important to clarify their formation mechanism. The liquid immersion method has a high potential for the elucidation of the compaction process. Recently, we have modified the liquid immersion method with a confocal laser-scanning microscope, CLSM.[2] In this method specimens are made transparent by an immersion liquid containing a fluorescent dye, and examined with a CLSM. This method is very promising for examining the change of internal structure in granule compact in the initial stage of compaction. An obstacle is to overcome the sample preparation, however. The mechanical strength is very weak in the tapped and/or pressed compact at very low pressure, and it is impossible to prepare thin specimens to observe the internal structure.

This problem can be solved with a novel method, a technique of impregnated resin coupled with the CLSM.[3] The method applies a resin of the same refractive index as the ceramic powder to provide the specimen both the mechanical strength and transparency. A small amount of fluorescent dye is added in the resin also. With the confocal optics, this technique provides very clear three-dimensional image of internal structure for all stages of compaction method.

2. Experimental

The starting material used in the present experiments is commercial alumina powder (AL160-SG1, Showa Denko Co. Ltd., Tokyo, Japan). The average particle size is 0.6 μm and the specific surface area 6.0 m^2/g. The starting material is mixed with distilled water in a ball-mill for 20 h. Small amounts of dispersant are used to prepare a flocculated slurry. The powder content in the slurry is 20 vol%. A spray dryer (Model No. SD-13, Mitsui Mining Co. Ltd., Japan) is used to prepare granule at inlet and outlet air temperatures of 200 °C and 100 °C, respectively. Sieving is used to prepare the granule of nominal size range 63–75 μm. The morphology of granules was examined with a scanning electron microscope (SEM, Model JSM-5310LV, JEOL, Tokyo, Japan).

The granules are placed in a transparent acrylic dies and pressed with the universal testing machine (Autograph AGI, Shimazu, Japan) at crosshead speed 0.5 mm/min after tapping. The tapped and/or pressed compacts are impregnated

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doi:10.1016/j.jeurceramsoc.2005.07.005

Fig. 1. Optical micrograph of a acrylic die filled with granules and impregnated resin. Arrows show a flow direction of resin.

by the resin by forcing the resin from the bottom of the die as shown in Fig. 1. The resin used in this study is MPV (bis(4-vinyl thio phenyl)sulfide, Sumitomo Seika Chemicals Co. Ltd., Osaka, Japan), which has a high refractive index ($n = 1.740$). A small amount of a fluorescence reagent (Nile red, Molecular Probe Inc., USA) is added in the resin. The compact impregnated with the resin is heated to 130 °C for curing. A sample is cut from the granule compact with resin, and is polished with an abrasive paper. A confocal laser-scanning microscope is used to observe the structure in the

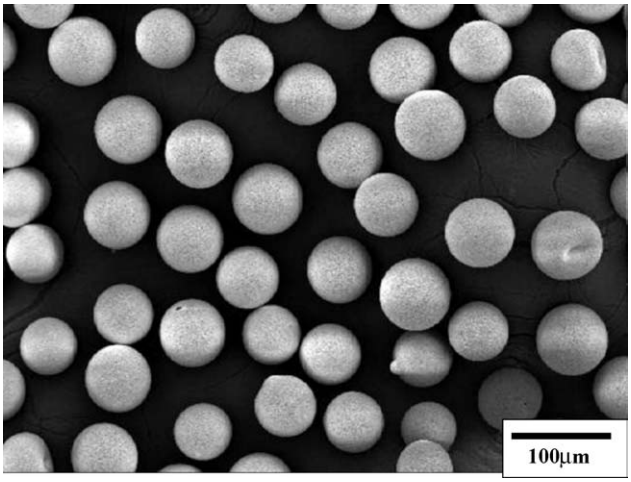

Fig. 2. SEM micrograph of sieved granules.

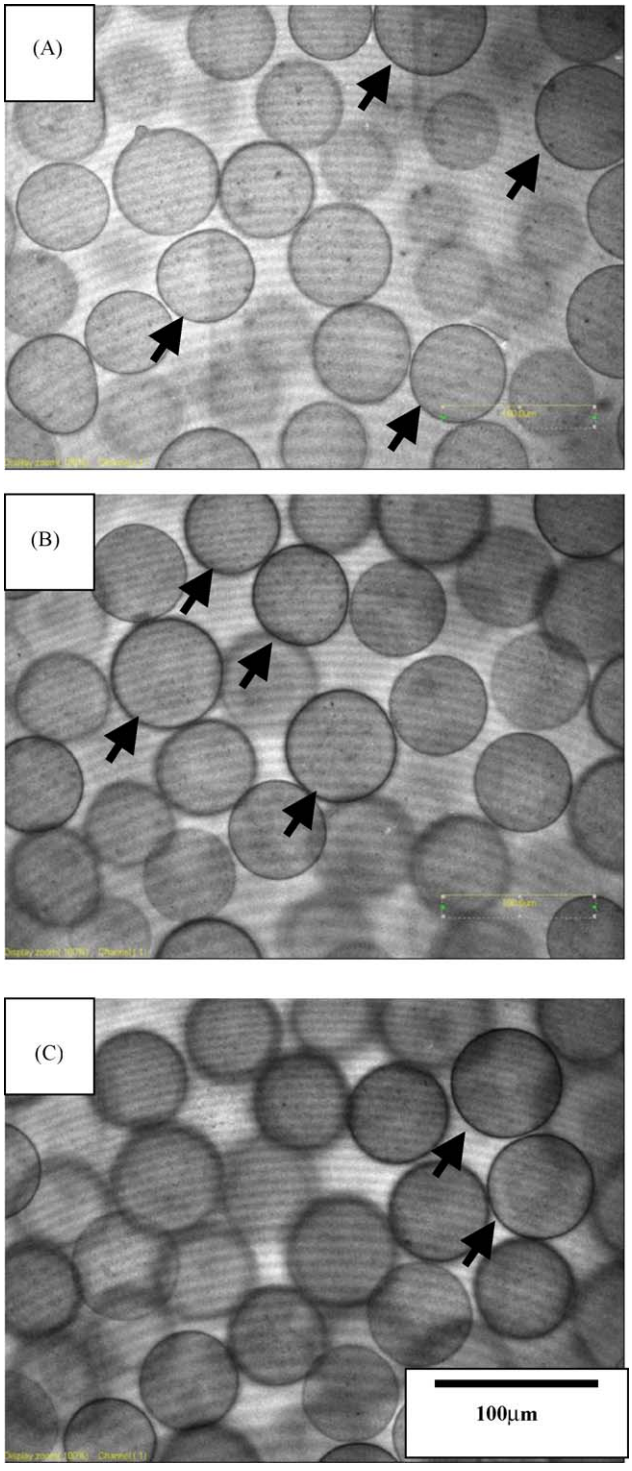

Fig. 3. CLSM images of internal structure of tapped granules taken at several depths: (A) surface, (B) 17 μm under and (C) 34 μm under. Arrows show outlines of granules in the focal plane.

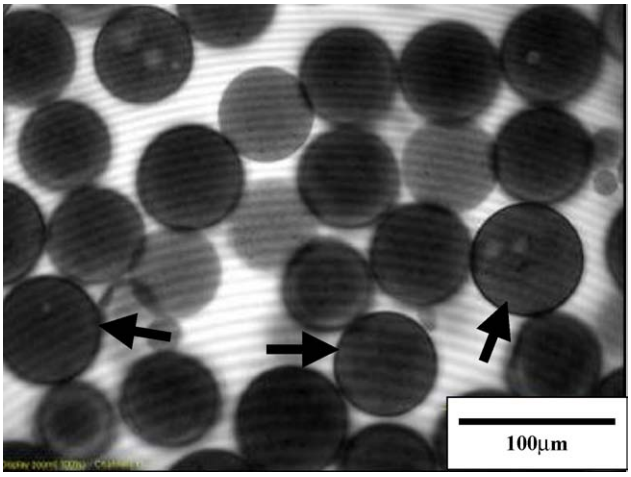

Fig. 4. CLSM images of internal structure of granules in the immersion liquid.

fluorescence mode (CLSM, FLUOVIEW, Olympus Co. Ltd., Japan).

3. Results and discussion

Fig. 2 shows SEM micrographs of the used granules. Granules prepared from flocculated slurry have spherical shapes. Their sizes are quite uniform. Clearly, the sieving ensured the uniform size.

Fig. 3 shows the CLSM images for the internal structure of tapped granules. In this mode of observation, the brightness of image corresponds to the porosity, i.e., open space appears bright and dense regions dark. The granules appear gray, since the space within the granules is half filled with the solid particles. These micrographs are taken by changing the specimen-to-lens distance at a fixed interval (10 μm) to examine the variation of structure with the depth in the die. It should be noted that this interval of observation is only apparent. The real interval of observation is 17 μm after correction with the refractive index of resin (1.74). Clear images are acquired for all places. The packing structure of the granules appears the same and very loose after tapping in the die. This result is reasonable, since the present microscopy shows the two-dimensional structure at a given plane. Only

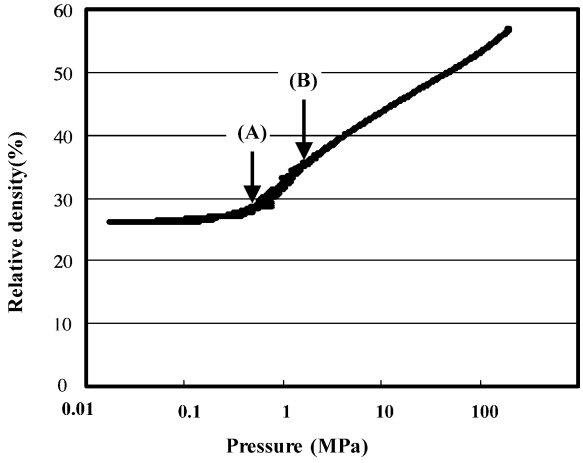

Fig. 5. Compaction curve of granules. Arrows A and B shows border of compaction stage.

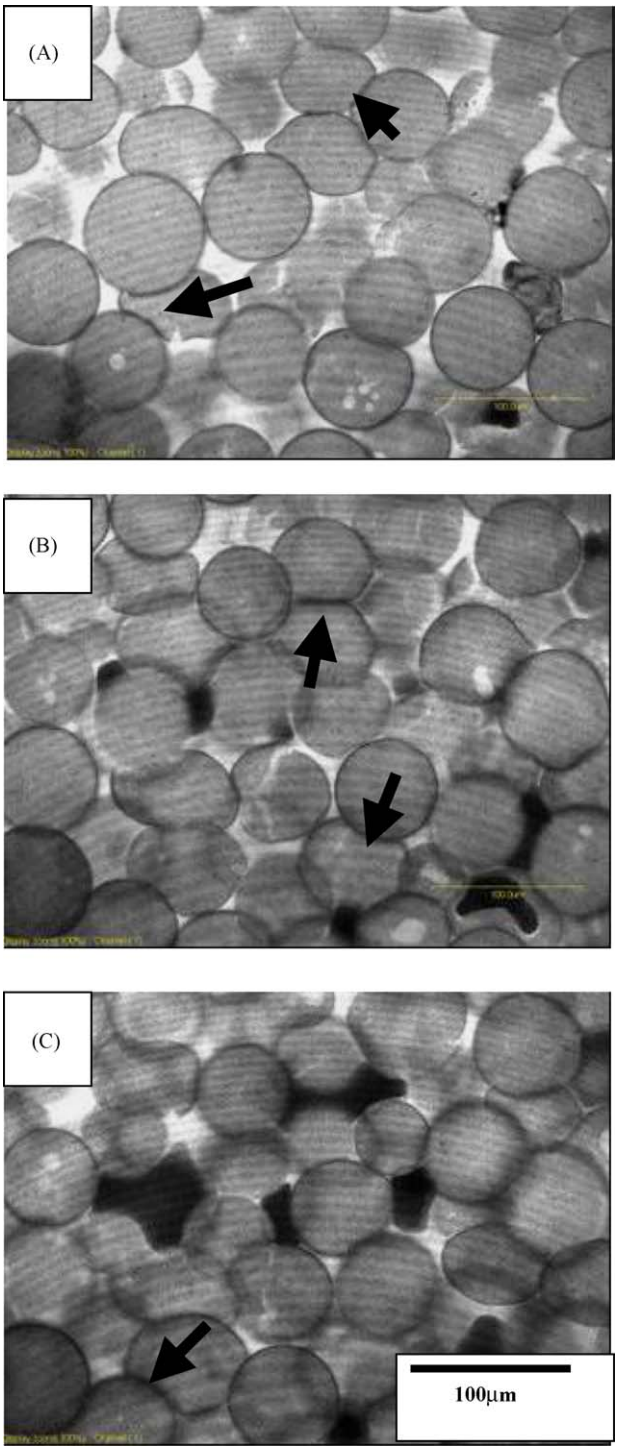

Fig. 6. CLSM images of internal structure of pressed granules at 2 MPa taken at several depths: (A) surface, (B) 17 μm under and (C) 34 μm under. Arrows show outline of deformed granules.

about one quarter of space is filled by the solid resulting in a packing density of 26% (about one-half of space by the granules of apparent relative density 50%). The optical image changed little also within the interval 34 μm (Fig. 3(A) and (C)), but the sharpness as shown by the arrows. Clearly, the resin can flow into the granules as well as the intergranular regions. The image obtained in this study is essentially the same to that taken with the conventional immersion liquid by a CLSM with a fluorescence mode as shown in Fig. 4.

Fig. 5 shows the compaction curve for die pressing of the granules. Three characteristic stages are noted in the compaction curve. The structure changes associated with these stages are explained as follows. Only the rearrangement of the granules is possible under the pressure 0.5 MPa (Fig. 5(A)). Deformation of the granules starts in the range 0.5–2 MPa (Fig. 5(B)), and the compact is densified by filling the intergranule space. No densification occurs within each granule at this stage. At high pressure, the spaces between the granules are filled, and the inter-particle distance in granules starts to decrease.[4] To verify this explanation, granules compacted at 2 MPa were solidified by the resin and the structure was examined by the new characterization technique.

Fig. 6 shows the CLSM images of internal structure of granules pressed at 2 MPa, which are taken at three depths in the compact. Arrows show the outline of deformed granules in three focal planes ((A)–(C)). The granules are much more densely packed than the tapped specimen of Fig. 3. Deformation of granules is noted at the region of granule contact. Surprisingly, however, many granules retain nearly the round shape. In addition, dark areas are noted in the image especially in the deep region, due to the absence of resin in

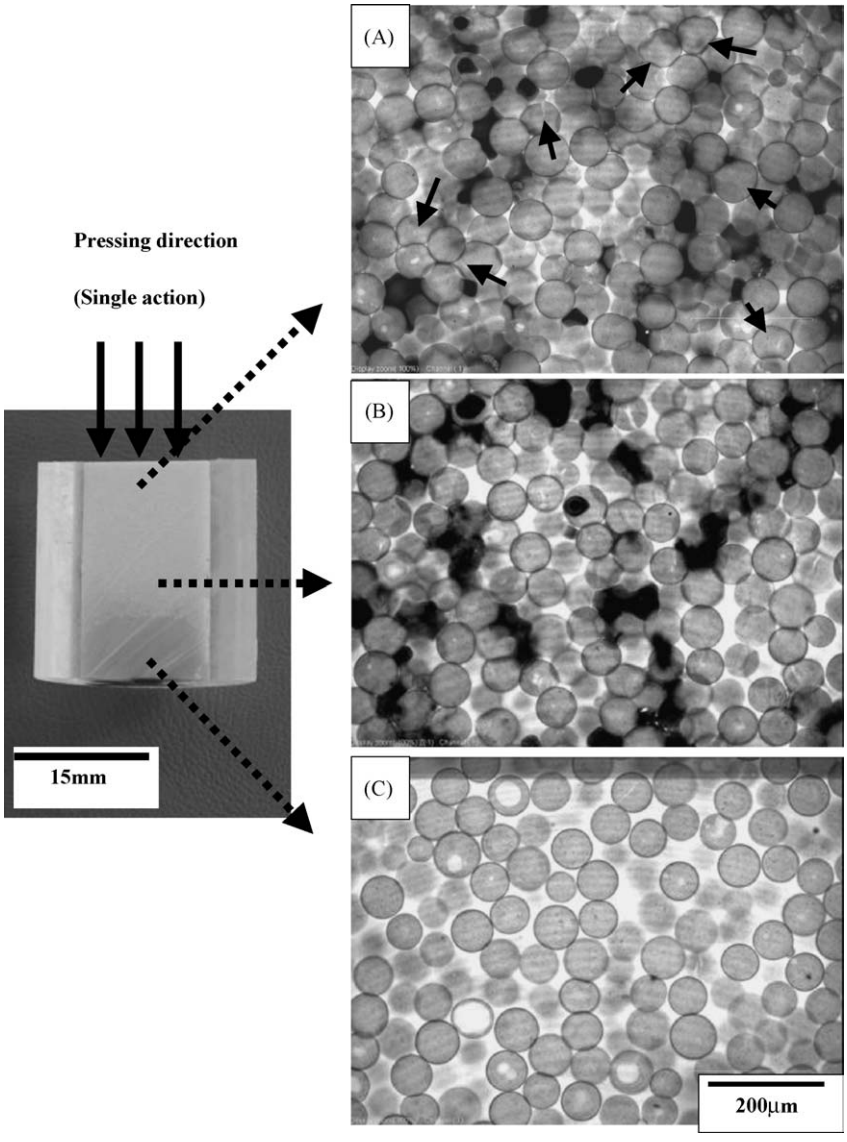

Fig. 7. Cross section of granule compact at 2 MPa of single action and CLSM image at different position in the compact: (A) top, (B) middle, (C) bottom. Arrows in the part (A) show outline of deformed granules.

these regions. Clearly, impregnation of resin is more difficult in this dense compact than the tapped granules of high porosity.

Fig. 7 shows the internal structures of a compact at various positions in the die. The structure is clearly different for these three locations. The deformation of granules decreases with increasing depth in the die. This is understandable. There is a pressure gradient in the compact in die pressing.[5] The pressure should be the highest at the top and decreases toward the bottom of the die. The present result is consistent to the recent study with the X-ray computed tomography (X-ray CT), in which the localized densification is noted during the compaction of alumina granules.[6] In concluding the study, it should be noted that the present results could not be obtained with conventional method such as SEM observation. It is impossible to prepare specimens for SEM, especially tapped specimens.[7]

4. Conclusion

A new characterization method using a resin of high refractive index is developed and applied for the first time in the direct study of powder compaction process. Clear images drastically deepened our understanding of structure during the compaction processing of granule. The changes of structure with location as well as the pressure were clearly understood. However, in the case of non-spherical granules, most commercial granules, it is difficult to discuss deformation process of granules quantitatively using this method.

Acknowledgements

We wish to thank Mr. K. Hamatani in Sumitomo Seika Chemicals Co. Ltd. in Japan for preparing the MPV, the high refractive index resin.

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